

The effect of fluctuations on the electrical transport behaviour in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$

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Received 15 April 1992, in final form 10 June 1992

Abstract. The excess conductivity behaviour of highly oriented $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films prepared by both coevaporation and laser ablation has been studied in detail in the reduced-temperature range $9 \times 10^{-4} < t < 1$. The excess conductivity in all the films studied was found to diverge sharply near T_c , in agreement with the conventional mean-field theory. However, the detailed temperature dependence could not be fitted to either the power-law or the logarithmic functional forms as predicted by the theory. The excess conductivity of all the films was found to be exponentially dependent on the temperature over nearly three decades for $9 \times 10^{-4} < t < 10^{-1}$, in contradiction to the mean-field theory.

1. Introduction

The rounding of the superconducting phase transition has been conventionally attributed to fluctuations in the magnitude and lifetime of the order parameter. By considering fluctuations of magnitude less than the order parameter in the Ginzburg-Landau (GL) theory, a critical temperature region was predicted in which the GL theory will not be valid. For temperatures greater than the critical limit the excess contribution $\Delta\sigma$ to electrical conductivity is estimated using the mean-field value of the order parameter in the time-dependent GL (TDGL) theory. The excess conductivity $\Delta\sigma$ is defined as $\sigma_{\text{exp}} - \sigma_{\text{calc}}$ where σ_{exp} is the experimentally observed electrical conductivity and σ_{calc} the conductivity due to normal-electron scattering alone in the absence of superconducting fluctuations. It is found to follow a power-law-type temperature dependence given by

$$\Delta\sigma(T) \propto (t)^{-\eta} \quad (1)$$

where t is the logarithm of the reduced temperature given by $\ln(T/T_c^{\text{mf}}) \simeq [(T - T_c^{\text{mf}})/T_c^{\text{mf}}]$ for $t < 1$ (T_c^{mf} is the mean-field transition temperature) and η a constant which depends on the dimensionality of conduction [1]. For three-dimensional (3D) conduction it is $\frac{1}{2}$ and for two-dimensional (2D) conduction it is 1. Hence the temperature dependence of $\Delta\sigma$ has been extensively studied in order to determine η and, thus, the dimensionality of order parameter fluctuations, the nature

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of contributions to excess conductivity and also to estimate the coherence length. In the case of $\text{YBa}_2\text{Cu}_3\text{O}_7$, equation (1) has been extensively used to determine the dimensionality of $\Delta\sigma$ in the range $10^{-3} < t < 1$ and it has been reported to be 3D, 2D and quasi-2D crossing over to a 3D behaviour close to T_c [2]. Recently, however, it was found that $\Delta\sigma$ does not exhibit the classical power-law dependence on t in the mean-field regime but instead has a logarithmic dependence [3]. The deviation from normal behaviour of the specific heat of an untwinned single crystal was also found to exhibit a logarithmic temperature dependence in the range $10^{-4} < t < 10^{-1}$ by Regan *et al* [4]. The contribution of fluctuations to diamagnetic susceptibility in bulk pellets on the other hand was found to obey the predictions of conventional GL theory [5]. Howson *et al* [6] have studied the variation in thermoelectric power up to T_c in single crystals and found that it exhibits an anomalous peak near T_c because of the presence of 3D divergent fluctuations. This large body of experimental results clearly indicates that the phenomenon of fluctuations and the length of the critical region in the oxide superconductors is still not completely understood.

In the present work we report the systematic study of $\Delta\sigma$ in highly oriented $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films prepared by two different techniques: coevaporation and laser ablation. The study of fluctuation effects requires the background or normal-state contribution to the overall conductivity to be accurately determined. Hence the normal-state behaviour was analysed in terms of both the linear metallic conduction phenomenon and the more recent resonating-valence-bond (RVB) model [7]. It was found that the film with the lowest room-temperature resistivity follows metallic conduction behaviour while the other films follow the RVB model. The temperature variation in $\Delta\sigma$ for all the films was found to deviate completely from that predicted by the conventional GL-based models.

2. Experimental methods

The thin films of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ were deposited onto SrTiO_3 (100) substrates by two methods: coevaporation and laser ablation. In the coevaporation, Y and Cu were electron beam evaporated while BaF_2 was resistively evaporated onto a cold substrate in an oxygen ambient. The thickness of the as-deposited film is $0.5 \mu\text{m}$. The films were later annealed at 850°C in wet flowing oxygen to form the superconducting phase. In the case of laser ablation the film ($0.3 \mu\text{m}$ thick) was deposited onto a heated substrate in an oxygen atmosphere from a sintered $\text{YBa}_2\text{Cu}_3\text{O}_7$ target. Structural characterization of the films was done by scanning electron microscopy and large-angle x-ray diffraction. The DC transport behaviour as a function of T was studied by the standard four-probe method. Thin Au wires attached to the film surface by In solder were used as leads for electrical characterization. These provided very-low-resistance ohmic contacts to the film surface. In the transition region, data points were taken at 0.2 K intervals to facilitate accurate analysis. The normal-state conductivity σ_{calc} for $T < 110 \text{ K}$ was determined by extrapolation of the regression-fitted data in the range $110 \text{ K} < T < 180 \text{ K}$.

3. Results

The microstructure of the two coevaporated films (C1 and C2) were completely different although they were deposited and annealed under apparently identical conditions.

Film C1 has long cylindrical grains of about $0.25\ \mu\text{m}$ diameter. The grains are highly oriented with their a - b plane along the film plane. Film C2, however, has a basket-weave-type grain morphology with an aspect ratio of about 16 in the film plane. The x-ray diffraction spectrum shows that the film has both c -axis- and a - b -axis-aligned grains along the film normal. The a - b -axis-aligned grains, however, were restricted to the top surface of the film [8]. The laser-ablated film (L) also shows a highly oriented cylindrical grain morphology similar to that of film C1. The film has a mirror-like smooth surface morphology, indicating that the surface roughness is much less than those of the coevaporated films.

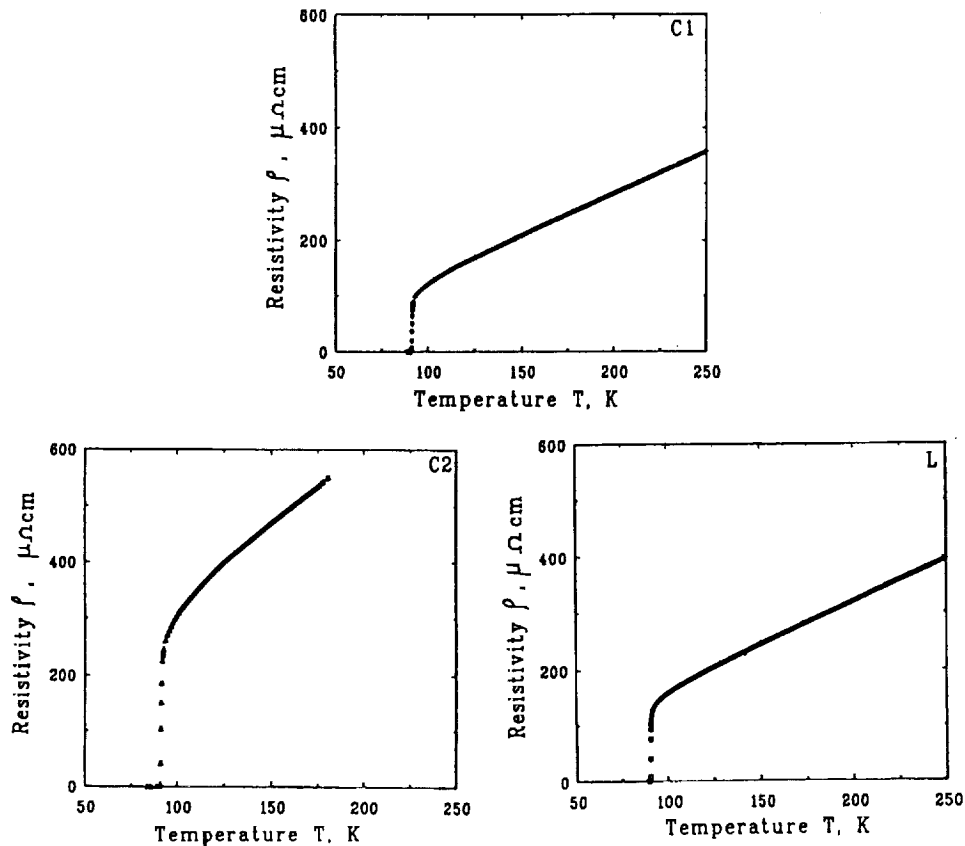


Figure 1. The variation in the resistivities ρ of the three films with temperature T .

The variation in the resistivities ρ with temperature of the three films C1, C2 and L shows a sharp transition into the superconducting state (figure 1). The transition width, defined as the width at half-maximum of the temperature-derivative curve, is about 0.4 K for all the films. The parameters that are important in any systematic study of $\Delta\sigma$ are

- (i) determination of the mean-field transition temperature T_c^{mf} and
- (ii) determination of the background or normal-state transport contribution to the overall conductivity.

It has been shown that the inflection temperature in the ρ - T curve can be

approximated as the mean-field transition temperature [9, 10]. This criterion was used in the determination of T_c^{mf} for the films and is given in table 1.

Table 1. The zero-resistance temperature $T_c(0)$, mean-field transition temperature T_c^{mf} and the transition width $\Delta T_c(0)$ to zero-resistance state of the three films.

Sample	$T_c(0)$ (K)	T_c^{mf} (K)	$\Delta T_c(0)$ (K)
C1	91.1	91.5	0.4
C2	90.5	91.4	0.5
L	90.2	90.5	0.3

4. Discussion

The low-temperature superconductors are of strong-coupling BCS type and their normal-state behaviour can be estimated using a linear temperature dependence for ρ based on the conventional electron scattering mechanisms. However, the scattering mechanisms in the normal state of the oxide superconductors are not clearly known. Both the Fermi-liquid-based models which predict a linear temperature dependence and the non-Fermi-liquid-based models are currently used to fit the normal-state transport behaviour [11]. According to the non-Fermi-liquid-based RVB model, the charge carriers are assumed to be confined to the Cu-O a - b planes of the crystal, thus leading to metallic conduction behaviour along the planes and phonon-activated hopping conduction in between the planes. The overall resistivity $\rho(T)$ in such an hypothesis is given by an expression of the form

$$\rho(T) = aT^{-1} + bT \quad (2)$$

where a and b are temperature-independent constants. In the present analysis, the ρ - T data of all the films were fitted to both the linear temperature dependence of the type $\rho(T) = \rho(0) + bT$ and the RVB dependence, equation (2), using the least-squares regression-fitting routine in the range $110 \text{ K} < T < 180 \text{ K}$. It was found that the best fit to film C1 was the linear relation while that for films C2 and L was the RVB-type relation. However, only the linear coefficient b of the two films C2 and L is in reasonable agreement with that predicted by the RVB model. The values of a in equation (2), 442.1 and 925.8 $\mu\Omega \text{ cm K}$, are orders of magnitude lower than the predicted values. The linear temperature coefficients b for the three films are 0.186 $\mu\Omega \text{ cm K}^{-1}$, 0.294 $\mu\Omega \text{ cm K}^{-1}$ and 1.584 $\mu\Omega \text{ cm K}^{-1}$, respectively. These values are well within the average values observed for single crystals [2, 3] and indicate the phase purity of the films. Using the simple Drude relation for metallic conduction given by $\rho(T) = (3\pi\hbar/2)/e^2k_F^2l$ where l is the quasi-particle mean free path, e the electron charge, \hbar the Planck constant and $k_F \simeq 4.46 \times 10^7 \text{ cm}^{-1}$ [12] the Fermi wavevector, the 'metallic parameter' $k_F l$ can be estimated for the three films. These were found to be 17, 7 and 14 for films C1, C2 and L, respectively, at 120 K, indicating that all the three films are very much on the metallic side of the Ioffe-Regel limit. This clearly shows that the microscopic conduction mechanism in these materials above T_c is not completely understood, although a mathematical fit

to the RVB model can be obtained. Recently, on the basis of mid-infrared phonon spectroscopy [13] and transport [14] studies on polycrystalline pellets it has been reported that the contribution from fluctuations persists up to temperatures as high as $2T_c$. This corresponds to the 'fluctuation onset' temperature, indicating that the lifetime of the superconducting fluctuations is finite and large even at $2T_c$, for which there is no direct experimental evidence at present.

The excess conductivity $\Delta\sigma(T) = \sigma_{\text{exp}}(T) - \sigma_{\text{calc}}(T)$ determined using the relation $\rho(T) = \rho(0) + bT$ for film C1 and equation (2) for films C2 and L was found to diverge sharply as T approaches T_c^{mf} . This is in qualitative agreement with the conventional theory which predicts a divergence of the magnitude of the order parameter fluctuations at T close to T_c^{mf} . The critical temperature region t in which the TDGL theory is not applicable can be estimated using typical values for $\text{YBa}_2\text{Cu}_3\text{O}_7$; $T_c^{\text{mf}} = 91$ K, the zero-temperature upper critical field $H_{c2}(0) = 674$ T and the GL parameter $K = 200$ [15], and therefore t is found to be about 2×10^{-2} . For $t \geq 2 \times 10^{-2}$, $\Delta\sigma(T)$ can in principle be determined using equation (1) (power-law dependence of $\Delta\sigma$ on t). Although the rate of decay of the fluctuating superconducting pairs is explicitly considered in obtaining equation (1), their effect on the quasi-particle conductivity is not considered. An additional term has been proposed to equation (1) by Maki and Thompson (as quoted by Skocpol and Tinkham [16]) to account for the effect of fluctuations on the quasi-particle conductivity and it was found to be four times equation (1) in the case of 3D conduction and $(e^2/8\hbar d)[(t - \delta)^{-1} \ln(t/\delta)^{-1}]$ for 2D conduction, where δ is the pair-breaking parameter and d the film thickness. The criterion for 2D conduction is $d/\xi(T) \ll 1$ where $\xi(T)$ is the superconducting coherence length. In the present case, even $d/\xi(0)$ for all the three films is much greater than unity and hence 2D conduction can be completely ruled out. The addition of an extra term to equation (1) changes only the magnitude of $\Delta\sigma(T)$, leaving the power-law temperature dependence intact. In the present work, however, $\Delta\sigma(T)$ for all the three films determined from the experimental data does not show a power-law dependence on t as predicted in the range $9 \times 10^{-4} < t < 4 \times 10^{-1}$; this can be clearly seen in figure 2. It has a continuously changing curvature which has been observed earlier. However, the previous reports have inferred changes in the dimensionality of electrical transport on the basis of linear fits to small portions of the curve [2].

The above method of analysis relies on the accurate determination of T_c^{mf} . In the case of oxide superconductors, the fluctuation effects on the conductivity are spread over a large temperature range compared with the conventional superconductors because of their extremely short coherence length ξ and the high value of $T_c(0)$. Hence the accurate determination of T_c^{mf} is difficult. An alternative method of analysing the fluctuation effects which does not depend on T_c^{mf} has been used in the case of Tl-Ba-Ca-Cu-O thin films and single crystals [17, 18]. According to this method, equation (1) can be rewritten as

$$[\Delta\sigma(T)]^{-1/\eta} = D^{-1/\eta} [(T - T_c^{\text{mf}})/T_c^{\text{mf}}] \quad (3)$$

where the constant D is given by $e^2/32\hbar\xi(0)$ for 3D conduction and $e^2/16\hbar d$ for 2D conduction. Differentiating and rearranging equation (3) gives

$$\ln[-d(\Delta\sigma)/dT] = \ln(D^{-1/\eta}/T_c) + (1 + 1/\eta) \ln(\Delta\sigma). \quad (4)$$

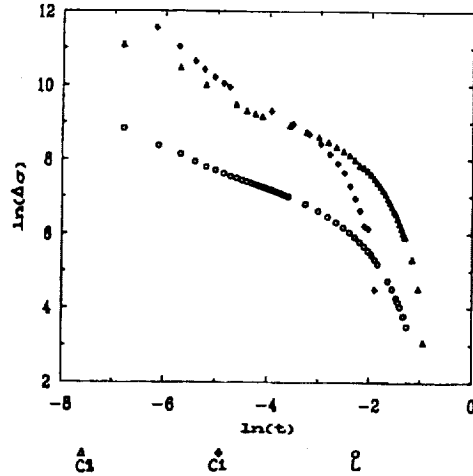


Figure 2. The excess conductivity $\Delta\sigma$ as a function of the reduced temperature $t = \ln(T/T_c^{mf})$ for all three films. It can be seen that a 'power-law' fit is not feasible except in small ranges of t . Δ , C2; \diamond , C1; \circ , L.

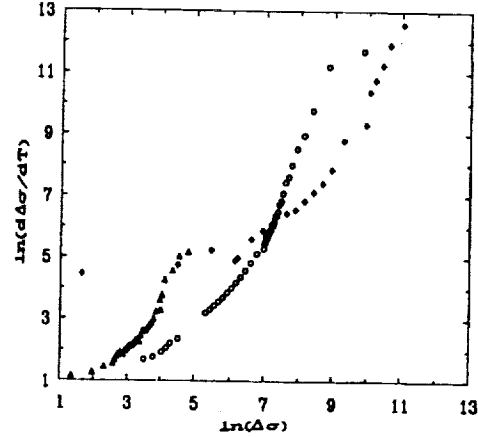


Figure 3. The excess conductivity $\Delta\sigma$ data represented according to equation (4), which is independent of T_c^{mf} . The presence of continuous curvature in the whole temperature range indicates a clear deviation from power-law behaviour.

The dimensionality η can be deduced from the slope of the $\ln[-d(\Delta\sigma)/dT]$ versus $\ln(\Delta\sigma)$ plot and using equation (4). Figure 3 shows $\Delta\sigma$ plotted according to this modified scheme. It can be clearly seen that even this alternative methodology which is independent of T_c^{mf} does not give conclusive evidence for the dimensionality of conduction in these films. Even according to this modified scheme of analysis the data exhibit a continuous curvature in the whole range and the dimensionality can be inferred only by fitting small portions of the excess conductivity. The recent electrical transport, mid-infrared phonon spectrum and heat capacity studies have clearly shown that the 'onset' temperature for fluctuations can be as high as about $2T_c$. The onset of fluctuations in the electrical transport behaviour has been attributed to the quasi-2D Maki-Thompson correction factor which has a logarithmic temperature dependence [3, 14]. The $\Delta\sigma$ -values in the present work, however, could not be fitted satisfactorily to a logarithmic temperature dependence. The $\Delta\sigma(T)$ data are replotted as shown in figure 4 and it can be clearly seen that $\Delta\sigma(T)$ has an exponential dependence on t : $\Delta\sigma(T) \propto \exp(t^{-\alpha})$ where α is the slope in the range $9 \times 10^{-4} < t < 10^{-1}$ for all the three films. This clearly illustrates two important points.

- (i) The TDGL theory underestimates the critical temperature region by at least an order of magnitude.
- (ii) The mean-field approximations are not valid in the case of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$.

The underestimation of the critical region has been attributed to the large contribution of the higher-order fluctuation corrections [19]. In the critical region, $\Delta\sigma$ is predicted to diverge as t goes to 0 with a temperature dependence similar to that in the mean-field region but with an exponent different from η based on the 3D XY model [20]. The results of the present work, however, cannot be understood even according to these models.

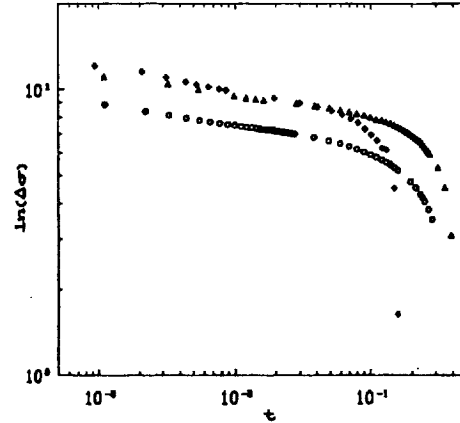


Figure 4. Log-normal plot of $\Delta\sigma$ versus t , showing the exponential temperature dependence of $\Delta\sigma$ for all the films in the range $9 \times 10^{-4} < t < 10^{-1}$.

5. Conclusions

The macroscopic and microscopic properties of the oxide materials in their superconducting state are being extensively studied. Many models have already been proposed to explain these properties. However, the normal-state behaviour remains the least studied to date. The only phenomenological model that has been proposed to explain the normal-state electron transport behaviour is the RVB model. The results of the present work indicate that it is insufficient to explain the transport behaviour above T_c . The linear and hopping coefficients obtained for the two films which obey the RVB expression for conductivity are much lower than the values predicted by the model.

The excess conductivity in the mean-field region of the three films studied does not obey the temperature dependence predicted by the TDGL theory. Even though such a behaviour has been observed before, the results are still fitted to the TDGL theory and the dimensionality of the electrical transport determined. However, we find that the excess conductivity is better represented by an exponential relation and that there is no model at present, macroscopic or microscopic, which can explain this type of behaviour. Hence the dimensionality of electrical transport is still inconclusive. The onset temperature for fluctuations observed in the present work, $t \simeq 0.1$, agrees with that observed by Regan *et al* [4], indicating that the length of the critical region is larger than that predicted by the theory. However, the functional dependence on temperature is found to be different.

Acknowledgments

SV wishes to acknowledge the US National Research Council for an Associateship at Lewis Research Center, NASA, and MAS acknowledges NASA for the support through grant NAG-440751. The authors acknowledge Professor J Scofield for the use of his evaporation system, J D Warner for the laser-ablated film, and T Harris and G Kitchen for their assistance. Critical reading of the manuscript by Dr Padma Satish is gratefully acknowledged.

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Ellipsometric study of ambient-produced overlayer growth rate on $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films

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(Received 28 August 1992; accepted for publication 18 January 1993)

An ellipsometric study of ambient-reaction-produced BaCO_3 overlayer growth on laser-ablated $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ is presented as a function of time. The effects of the anisotropy of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ on the ellipsometric data inversion process are discussed, and it is concluded that with certain restrictions on the data acquisition method, the anisotropic substrate can be adequately modeled by its isotropic pseudodielectric function for the purpose of overlayer thickness estimation. It is found that after an initial period of rapid growth attributed to the chemical reaction of the exposed surface bonds, the BaCO_3 overlayer growth is linear at 1–2 Å per day. This slow growth rate is attributed to the complexity of the BaCO_3 forming reaction, together with the need for ambient reactants to diffuse through the overlayer.

I. INTRODUCTION

Previously,¹ we have reported results of ellipsometric measurements of the pseudodielectric function of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) prepared by laser ablation and co-evaporation. We also reported observing growth of a transparent overlayer on the laser-ablated films. This overlayer growth has been observed by other experimenters² and has been determined to be BaCO_3 resulting from interaction between YBCO and CO_2 in humid air,³⁻⁵ a conclusion with which we concurred. In this article we report systematic ellipsometric measurements of the growth rate of this overlayer on laser-ablated films exposed to air. We discuss the effects of the anisotropy of the YBCO substrate on the ellipsometric inversion process and show that by fixing certain ellipsometer settings the effects of the anisotropy can be minimized. We further show that under these restrictions the YBCO substrate can be approximated by its isotropic pseudodielectric function for the purpose of estimating the transparent overlayer thickness. Finally, we present the results of the overlayer growth time dependence measurements and discuss the results in light of the chemical mechanism which is believed to create the overlayer.

II. EXPERIMENT

Samples were prepared by laser ablation¹ using an excimer laser operating at 248 nm, energy density of 1.5 J/cm²/pulse, with 4 pulses per second. The target was a sintered 25-mm-diam YBCO pellet located 8 cm from the sample at 45° to the laser beam. The beam was rastered up and down 1 cm over the target using an external lens on a translator. The films were deposited on strontium titanate (SrTiO_3), zirconium dioxide (ZrO_2), and lanthanum aluminate (LaAlO_3) substrates, which were mounted on a stainless-steel plate heated to 775 °C. The oxygen pressure was 170 mTorr throughout the deposition. X-ray diffraction showed the films to be *c*-axis aligned. Comparison¹ of our measured pseudodielectric functions with published

data⁶ also indicated our films closely approximated *c*-axis-aligned YBCO single-crystal material. Critical temperatures for the films were ~86 K. Film thicknesses averaged about 3000 Å.

The rotating analyzer spectroscopic ellipsometer system⁷ reflects monochromatic linearly polarized light off the sample and measures the complex reflection ratio ρ :

$$\rho = \frac{E_{p,r}/E_{s,r}}{E_{p,i}/E_{s,i}} = \left(\frac{E_{p,r}}{E_{s,r}} \right) \left(\frac{E_{s,i}}{E_{p,i}} \right) = \frac{E_{p,r}}{E_{s,r}} \tan(\theta), \quad (1)$$

where $E_{p,r}$ and $E_{s,r}$ are the parallel and perpendicular components (with respect to the plane of incidence) of the reflected electric field intensity, $E_{p,i}$ and $E_{s,i}$ are the corresponding quantities for the incident light, and θ is the incident light polarization azimuth, i.e., $\tan(\theta) = E_{s,i}/E_{p,i}$. The rotating analyzer ellipsometer actually measures the $E_{p,r}/E_{s,r}$ ratio, while the $E_{s,i}/E_{p,i}$ ratio is determined by a fixed polarizer azimuth θ . Two films, identified here as samples A and B, both deposited on strontium titanate, were selected for systematic monitoring of overlayer growth. These samples were cleaned with a bromine etch (1% Br/ethanol solution for 30 s followed by ethanol and blow drying)⁵ to remove the overlayer, and were measured less than 10 min after cleaning, and periodically thereafter. Both samples were left mounted throughout the growth monitoring period to maximize precision. The samples were monitored for a period of 4–10 days after cleaning, and were exposed to the air throughout the monitoring period. Each sample was etched two different times to determine repeatability, giving four etches in all. Additionally, sample A was mechanically cleaned and measured periodically over a period of 106 days to examine longer-term overlayer growth. Here the sample was not left mounted continuously but rather mounted periodically for measurements; hence the precision is poorer for this long-term monitoring. Between measurements, the sample was left exposed to air. Overlayer growth measurements were taken at a fixed angle of incidence: 65° for the first etch of

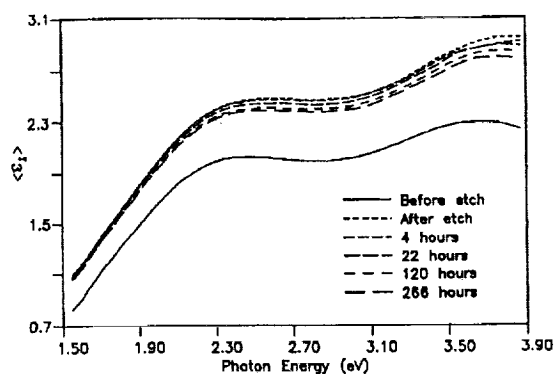


FIG. 1. Experimental $\langle \epsilon_1 \rangle$ for several representative scans of sample A taken before and after the first etch.

each sample and 70° for the second etch and also for the long-term monitoring of sample A; the wavelength range varied slightly but was always within the range 3200–8000 Å (1.55–3.87 eV). The polarizer azimuth θ was held constant at 24° for the first etches and also for the long-term monitoring of sample A; 20° was used for the second etch of each sample. These polarizer values have been shown to provide maximum precision, i.e., $|\rho| \approx \tan(\theta)$.⁸

III. RESULTS

Figure 1 shows $\langle \epsilon_1 \rangle$, the real part of the pseudodielectric function, for several representative measurements taken before and after the first etch of sample A. The pseudodielectric function is the apparent dielectric function of the sample, i.e., the dielectric function of an equivalent isotropic bulk material calculated using an isotropic two-phase (ambient/substrate) model.⁹ The pseudodielectric function $\langle \epsilon \rangle$ is obtained directly from the ellipsometrically measured complex reflectance ratio ρ , using the formula⁹

$$\langle \epsilon \rangle = \epsilon_a \left[\sin^2(\phi) + \sin^2(\phi) \tan^2(\phi) \left(\frac{1-\rho}{1+\rho} \right)^2 \right] \quad (2)$$

In the above equation ϵ_a is the dielectric function of the ambient ($\epsilon_a=1$ for air), ϕ is the experimental angle of incidence, and ρ is the ellipsometrically measured quantity defined previously. For an ideal two-phase system, $\langle \epsilon \rangle = \epsilon_n$, the substrate dielectric function. However, (2) defines the pseudodielectric function $\langle \epsilon \rangle$ for all cases. The lowest curve in Fig. 1 is the measurement taken prior to the bromine etch. The highest curve is the measurement taken immediately after the etch. The curves in between are measurements taken at later times. The magnitude of $\langle \epsilon_1 \rangle$ decreases with increasing time. The measured quantity $\langle \epsilon \rangle$ is expected to be a continuous function of overlayer thickness, and computer simulation using an isotropic substrate whose pseudodielectric function was similar to our measure $\langle \epsilon \rangle$ (Ref. 6) showed that the magnitude of the real part of the pseudodielectric function $\langle \epsilon_1 \rangle$ in our range of measurement decreased with increasing overlayer thickness, as shown in Fig. 2. This is in accordance with our

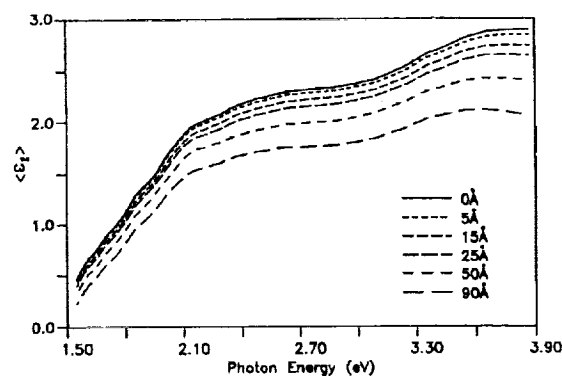


FIG. 2. Computer simulation of overlayer ($n=1.55$, $k=0$) growth on anisotropic substrate. $\langle \epsilon_1 \rangle$ is calculated at angle of incidence 70° . The curve labeled 0 Å corresponds to the actual substrate spectrum, which is taken from Ref. 6.

measurements: The large increase in $\langle \epsilon_1 \rangle$ magnitude immediately after etching indicates removal of the overlayer, and the steady decrease in magnitude as time increases indicates overlayer growth. Similar results are seen for the other growth monitoring measurements. Figure 3 shows the same information for the second etch of sample B in a different form. Here the real part of the pseudorefractive index $\langle n \rangle$ ($\langle \epsilon \rangle = \langle n \rangle^2$) at incident light wavelength of 4900 Å is plotted as a function of time. Again, a steady decrease in the magnitude of the real part with increasing time is seen. This effect of a transparent overlayer on the real part of $\langle n \rangle$ was also verified by computer simulation. Both the actual measurement of $\langle \epsilon_1 \rangle$ and the computer simulations of $\langle \epsilon_1 \rangle$ show that the effect of the overlayer is much more pronounced at higher photon energies (lower wavelengths). This is because the low-energy light penetrates deeper into the substrate, making it less sensitive to the overlayer.

IV. DATA ANALYSIS AND DISCUSSION

From the pseudodielectric function of single-crystal YBCO (Ref. 6) we estimate the light penetration depth in the range of measurement to be under 750 Å, making the

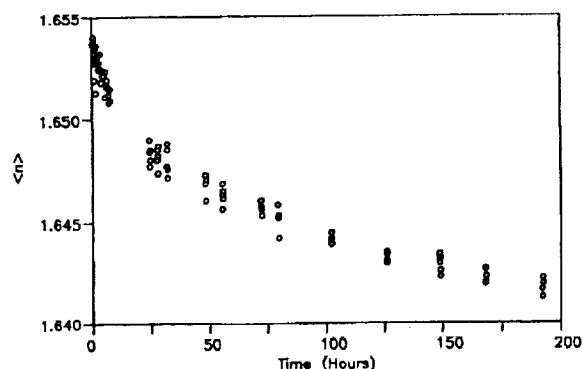


FIG. 3. Measured real part of $\langle n \rangle$ at light wavelength 4900 Å as a function of time for sample B, second etch.

YBCO film the effective substrate for ellipsometric purposes. To obtain estimates of the overlayer thicknesses, it is necessary to invert the ellipsometric data. Because of the mathematical complexity of this inversion process, a least-squares fit is usually performed.¹⁰ A model of the system must be formulated, and appropriate model parameters optimized in the least-squares sense with respect to the measurement. Such a model typically consists of a layered structure atop an optically thick substrate. Abrupt interfaces are assumed, and layers are assumed to be isotropic and homogeneous with regard to thickness and optical properties.¹¹ For overlayer measurements, the pseudodielectric function of the specific substrate to be studied is typically measured prior to overlayer deposition after sufficient cleaning to approximate the two-phase model. The overlayer is then introduced and the sample remeasured and analyzed with a three-phase (ambient/overlayer/substrate) model, using the pseudodielectric function of the initial uncontaminated surface measurement to model the substrate optical properties. This process provides a built-in correction for imperfections in the substrate which would not be accounted for by the use of standard reference data.⁹ Additionally, if the sample is left mounted on the ellipsometer between initial measurement of the substrate pseudodielectric function and subsequent measurement of the three-phase system, as was done in this study, effects of substrate inhomogeneity are also minimized by guaranteeing that the optical constants used to model the substrate are optimum for the specific location at which overlayer measurements are being taken.

In the present case, the above procedure is complicated by the fact that YBCO is a biaxially anisotropic material.¹² The pseudodielectric function is calculated assuming an isotropic two-phase system, and thus it cannot completely describe the optical properties of the anisotropic substrate, which should be described by a dielectric tensor.¹² To investigate the possible effects of this anisotropic substrate on the measurement of a transparent overlayer, we took two approaches: First, we performed computer simulations of untwinned single-crystal YBCO to determine which system parameters are affected by the anisotropy and to what degree. This provided information on how best to set up the overlayer measurement to minimize potential inaccuracies caused by the anisotropy, as well as providing bounds on the anisotropy-induced error. Second, we performed experimental measurements to identify to what degree the anisotropy effects predicted by computer simulation for untwinned single-crystal YBCO were actually seen in the laser-ablated YBCO films. These films are not single crystals but rather consist of microscopic grains of *c*-axis-oriented YBCO crystals with the *a* and *b* crystal directions aligned with the strontium titanate substrate crystal directions.^{2,12} There will be a high degree of twinning in the *a*-*b* plane.^{2,12} We expect this type of crystal structure to reduce the observable effects of the anisotropy.

The computer simulations of ellipsometric measurements of untwinned single-crystal YBCO were based upon the biaxial substrate model developed by Graves.^{11,13} This model calculates the complex amplitude reflection coeffi-

cients for a biaxially anisotropic substrate oriented such that one crystal axis is perpendicular to the sample surface while the a second crystal axis is perpendicular to the plane of incidence of the light. In the present case, the YBCO films are predominantly *c*-axis aligned, so that the *c* axis is perpendicular to the sample surface as required by the Graves model. The dielectric tensor components of YBCO were taken from Kircher *et al.*¹² Simulations were done using the *c* axis perpendicular to the interface and either the *b* axis perpendicular to the plane of incidence (*abc* orientation) or the *a* axis perpendicular to the plane of incidence (*bac* orientation). Physically this corresponds to measurement of an untwinned single crystal of YBCO, for which the maximum observable anisotropy effect would be expected. Comparison of the pseudodielectric function of *abc*-oriented simulations with the pseudodielectric function of *bac*-oriented simulations shows the effect of a 90° rotation on the measurement of such a single crystal. We performed these simulations at an angle of incidence of 65°, and found that the difference between the *abc*-oriented crystal pseudodielectric function and the *bac*-oriented crystal pseudodielectric function is enormous, with differences between the real and imaginary parts of $\langle \epsilon \rangle$ of the two orientations exceeding the absolute magnitude of these quantities. For example, at the wavelength 5500 Å, $\langle \epsilon_1 \rangle = 3$ for the *abc* orientation, while $\langle \epsilon_1 \rangle = 1$ for the *bac* orientation. Additionally, the shape of the two spectra differed significantly. In contrast, our measurements of laser-ablated YBCO films as a function of sample azimuth showed variation in $\langle \epsilon_1 \rangle$ and $\langle \epsilon_2 \rangle$ of less than 0.3 in all cases, and the shape of the spectra measured at different sample azimuths was very similar. The dependence of the ellipsometric measurement on sample azimuth has been reduced significantly by the complex crystal structure of the laser-ablated films, but it is still detectable, as shown by these measurements.

We next simulated the pseudodielectric function of untwinned single-crystal YBCO with no overlayer at various angles of incidence. Our simulations showed that the pseudodielectric function is a strong function of the ellipsometer angle of incidence for both the *abc* orientation and the *bac* orientation, with variation in spectral magnitudes greater than 15% for incident angle variation from 65° to 75°. The variation of the pseudodielectric function for each orientation was in opposition: the real part of $\langle n \rangle$ decreased with increasing angle of incidence for the *abc* orientation, whereas it increased with increasing angle of incidence for the *bac* orientation. For comparison, we measured numerous films at various angles of incidence. The measured pseudodielectric function was found to vary by 5% or less. Again, the effect of the anisotropy has been greatly reduced by the complex film structure, but has not been completely eliminated.

In our third simulation, we determined bounds on the error that can be expected in using an isotropic three-phase model to analyze a system with an anisotropic substrate. Using the anisotropic model, we simulated ellipsometric data for various overlayer thicknesses (overlayer $n = 1.55$, $k = 0$) at an angle of incidence of 70°. This generated data

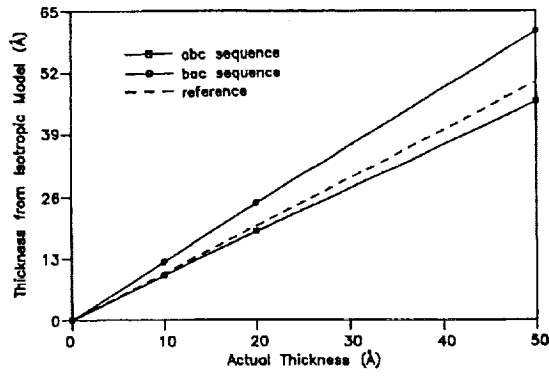


FIG. 4. Computer simulation using a three-phase isotropic model to analyze a system with an anisotropic substrate. *abc*: *c* axis perpendicular to interface, *b* axis perpendicular to plane of incidence; *bac*: *c* axis perpendicular to interface, *a* axis perpendicular to plane of incidence. Reference is calculated isotropic thickness equal actual thickness.

was then inverted using the isotropic three-phase model to obtain the overlayer thickness, using the simulated pseudodielectric function of the anisotropic substrate with no overlayer as the effective isotropic substrate. Simulation results are given in Fig. 4. The reference line in this graph is the ideal result, i.e., the overlayer thickness determined by the isotropic model exactly equals the overlayer thickness simulated on the anisotropic substrate. The absolute error is 20% or less, and the calculated thickness is directly proportional to the actual thickness. Thus, use of the isotropic model in this case may result in some error in the absolute growth rate, but the shape of the growth curve will be correct. As with the angle-of-incidence variation, the effects of the two orientations *abc* and *bac* oppose each other, with the *abc*-oriented crystal resulting in an underestimated thickness while the *bac*-oriented crystal results in an overestimate. In view of our previous results showing significant reduction in the observed effect of YBCO anisotropy in the laser-ablated films, the boundaries shown in Fig. 4 are expected to greatly overestimate the actual overlayer measurement error.

One potential effect of substrate anisotropy on ellipsometric measurements that cannot be studied using the Graves model is the effect of off-diagonal components of the reflectance matrix which result when the crystallographic axes are not aligned with the optical axes as required by Graves model. In general, the relationship between the incidence electric-field vector and the reflected electric-field vector is given by¹¹

$$\begin{bmatrix} E_{p,r} \\ E_{s,r} \end{bmatrix} = \begin{bmatrix} R_{pp} & R_{ps} \\ R_{sp} & R_{ss} \end{bmatrix} \begin{bmatrix} E_{p,i} \\ E_{s,i} \end{bmatrix} \quad (3)$$

Equation (3) states that the reflected complex amplitudes are related to the incident complex amplitudes by a 2×2 reflection matrix. This matrix is a property of the sample and is a function only of wavelength and angle of incidence. For either an isotropic system or a biaxially anisotropic system with the axial alignment specified for Graves model $R_{ps} = R_{sp} = 0$ and the ellipsometrically measured ra-

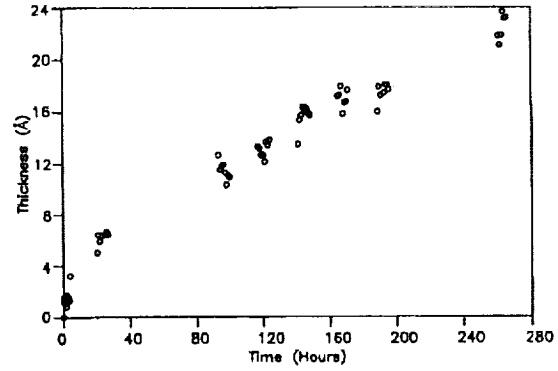


FIG. 5. Results of isotropic three-phase modeling of sample A, first etch growth monitoring. The plotted overlayer thickness is the only model parameter.

tio ρ reduces to $\rho = R_{pp}/R_{ss}$.¹¹ Thus, ρ is a function only of the reflection matrix, which is a property of the sample. In the case of a biaxially anisotropic system in which the optical axes and crystallographic axes do not coincide, R_{sp} and R_{ps} are not generally zero. The measured ratio ρ can be written as

$$\rho = \left(\frac{E_{p,r}}{E_{s,r}} \right) \left(\frac{E_{s,i}}{E_{p,i}} \right) = \frac{R_{pp} + R_{ps} \tan(\theta)}{R_{sp} + R_{ss} \tan(\theta)} \tan(\theta). \quad (4)$$

The ellipsometric measurement in this case is a function of both the reflectance matrix and the polarizer azimuth θ . We measured the effect of the polarizer azimuth setting both on a laser-ablated YBCO film and also on an isotropic reference sample. The reference sample was an amorphous carbon film on silicon, similar to samples described in Ref. 7. The film was nearly transparent ($k < 0.13$) with thickness ~ 1950 Å. For such a sample the amplitude and phase of ρ oscillates slowly (one complete cycle in our spectral range), so that we could locate spectral regions where the measured ρ of the YBCO film could be compared with measured ρ values of similar magnitude and phase for an isotropic system. The polarizer azimuth was varied between 20° and 70° . In this range, the variations of ρ for the isotropic sample are within the experimental error. For the YBCO film the variations in $\langle \epsilon_1 \rangle$ and $\langle \epsilon_2 \rangle$ are less than 8% of the amplitude between polarizer values of 20° and 70° . Between 20° and 45° the changes are less than 4%. Again, a small but observable effect of the anisotropy is seen in the laser-ablated film.

Based upon the above results, we used the measurement and analysis procedure outlined previously for isotropic systems, with the additional constraints of using a fixed angle of incidence and a fixed polarizer azimuth, so as to avoid any measurement variations not directly attributable to surface changes. Also, since each sample was left mounted throughout the overlayer growth measurement, the sample azimuth remained unchanged. The overlayer was modeled as a transparent dielectric material with refractive index of 1.55,¹ and the YBCO substrate was modeled by the pseudodielectric function measured immediately after etching. The resultant overlayer thickness

TABLE I. Results of linear regression analysis applied to growth monitoring results analyzed by an isotropic three-phase model. Only measurements taken after 12 h or longer were included in the fit. R is the correlation coefficient; $R = \pm 1$ is a perfect fit. m denotes mechanically cleaned.

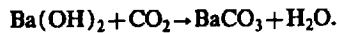
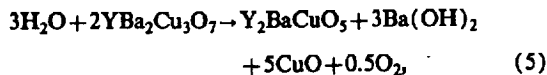
Sample	Etch	Hours	Rate ^a	Intercept ^b	R
A	1	266	1.62	5.03	0.983
A	2	95	2.12	6.62	0.983
A	m	2557	0.95	2.29	0.981
B	1	214	2.01	1.80	0.985
B	2	192	0.87	6.42	0.937

^aThe rate in Å/day.

^bThe intercept in Å.

versus time plot for the first etch of sample A is given in Fig. 5. Two distinct regions are seen. For about the first 5 h the overlayer grows rapidly. When the overlayer reaches ~ 5 Å the growth slows and becomes linear, with a growth rate of 1.62 Å per day, as determined by linear regression. Results for the other three etches were similar in shape. The four etch results plus the long-term monitoring of sample A after mechanical cleaning were each fitted to a linear model for overlayer measurements taken after 12 h, i.e., in the linear region. The results are shown in Table I. In this table, the intercept is the y intercept of the regression line; this gives an estimate of the thickness reached in the initial rapid growth region. The initial growth period stops at about 2–6 Å, i.e., about one monolayer. In the linear region, the growth rate is in the range 1–2 Å per day, which is below one monolayer per day.

The type of growth curve observed here on YBCO is different from the logarithmic curve typically observed in the oxidation of semiconductors, such as the oxidation rate of silicon and gallium arsenide measured by Lukeš.¹⁴ In the case of oxidation, the ambient reactant O_2 is plentiful, and the overlayer growth rate is controlled by the diffusion rate of oxygen through the growing oxide overlayer. In the present case, the YBCO overlayer is believed to be produced by the following chemical reactions:³



These reactions are clearly more complicated than a simple oxidation reaction, and also involve two reactants, H_2O and CO_2 , which make up a much smaller molar fraction of air than does O_2 . The growth curve observed here is quite similar to the growth rate of silver sulfide tarnish on silver exposed to room air,¹¹ where an initial period of rapid growth is seen up to 2 Å, after which the growth rate slows and becomes linear at ~ 4 Å per day.

Variations in the ultimate YBCO overlayer thickness reached during the initial rapid growth phase are probably due to two factors: variations in surface quality, and dif-

ferences in the delay time between etching and the initial measurement of the substrate dielectric function. This latter factor will be particularly significant if the initial growth curve shape is logarithmic; any overlayer growth that occurs prior to the initial measurement will be absorbed into the effective substrate measurement. Variations in the linear region slope may be due to variations in atmospheric conditions such as humidity which would affect the availability of the limiting reagents in Eq. (5). As a final comment, we note that the final measurement at 2557 h for the mechanically cleaned long-term monitoring of sample A gave a thickness of 100 Å as analyzed by the isotropic three-phase model. This measurement is the measurement labeled "Before etch" on Fig. 1; the effect of the removal of this 100 Å overlayer by the bromine etch is seen very clearly in the pseudodielectric function.

V. CONCLUSIONS

We have measured ellipsometrically insulator overlayer growth on laser-ablated YBCO thin films due to exposure to air. After formation of an initial monolayer, the growth proceeds linearly and rather slowly, e.g., 1–2 Å per day. This information should be useful in appraising the effect of air exposure on various sample processing steps, such as making electrical contacts. In the process, we considered the effects of substrate anisotropy on overlayer estimation and have presented a method of estimating overlayer growth on an anisotropic substrate.

ACKNOWLEDGMENTS

The authors would like to thank Jill E. Dunning for help in some of the growth monitoring measurements, and Dr. J. Kircher for supplying a diskette of the YBCO tensor component spectra.

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Magnetic flux relaxation in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin film: thermal or athermal*

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(Received March 15, 1991; revised May 28, 1991)

Abstract

The magnetic flux relaxation behavior of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin film on LaAlO_3 for $H \parallel c$ was studied in the range 4.2–40 K and 0.2–1.0 T. Both the normalized flux relaxation rate S and the flux pinning energy U_0 exhibit a weak field dependence at low temperatures ($T \leq 20$ K). Within this regime S and U_0 are observed to increase continuously from 1.0×10^{-2} to 2.0×10^{-2} and 45 to 130 meV respectively, as the temperature T increases from 4.2 to 20 K. While S is observed to decrease in proportion to kT for $T \leq 20$ K, it does not extrapolate to zero at $T = 0$, which is in contradiction to the thermally activated flux creep and vortex glass models. This behavior is discussed in terms of the athermal quantum tunneling of flux lines. The magnetic field dependence of U_0 , however, is not completely understood.

1. Introduction

In type II superconductors the pinning of magnetic flux lines is responsible for the lack of dissipation during the flow of high current densities. The pinning is caused by various types of defects, *i.e.* grain boundaries, twins, point defects and inhomogeneities. The observation of a high degree of mobility of these flux lines in the oxide superconductors [1–3] has stimulated many theoretical studies and has led to the proposal of several models. According to the conventional thermal flux motion model [4, 5] the magnetization relaxes logarithmically with time t for $t < t_{cr}$, where t_{cr} is a crossover time given by $t_{cr} = t_{hop} \exp[U/kT]$, t_{hop} is the flux line hopping time (10^{-6} – 10^{-12} s), U is the net flux pinning energy, k is the Boltzmann constant and T is temperature. At $t > t_{cr}$, or for high T , the motion of flux lines attains a steady state and the magnetization relaxes exponentially with t [6]. However, the observation of logarithmic decay even at large values of T and the non-linear behavior of the relaxation rate has led to

many alternative models for the nature of the pinning energy U [7–9].

An alternative description of dissipation in high temperature superconductors is the vortex glass model [10, 11]. In this model there is a truly superconducting state in the presence of high magnetic fields below the glass transition temperature. Within this regime the sample voltage is predicted to vanish exponentially with decreasing current. Recently, the vortex glass model has been used to explain the apparent temperature independent value of $S = 0.02$ – 0.035 reported by many researchers [12].

In the present work, the relaxation of screening-current-induced magnetization in a $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin film has been studied as a function of temperature T and external field H . The magnetization is found to relax logarithmically up to 10^4 s for T as high as $0.45 T_c$, where T_c is the superconducting transition temperature. S appears to saturate with increasing temperatures for $T > 20$ K. This observation is consistent with the predictions of the vortex glass model; however the $[\ln(t)]^{-1}$ time dependent relaxation appropriate to the model is not observed. For $T \leq 20$ K, S is observed to decrease linearly with decreasing T but does not extrapolate to zero at $T = 0$. The linear dependence of S on T in the low- T region is consistent with both the thermally activated creep and vortex glass models. The finite value of S at $T = 0$ obtained by extrapolation cannot be explained by either model and is discussed in terms of “quantum tunneling” or the “athermal flux motion” model [13]. We also observe U_0 to decrease with increasing H for all temperatures and fields used,

*Presented at the 18th International Conference on Metallurgical Coatings and Thin Films, San Diego, CA, USA, April 22–26, 1991. In error, an unrevised version of this paper was published in *Thin Solid Films*, 206 (1991) 137–142.

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although the $1/H$ dependence suggested by Yeshurun and Malozemoff [1] is not observed.

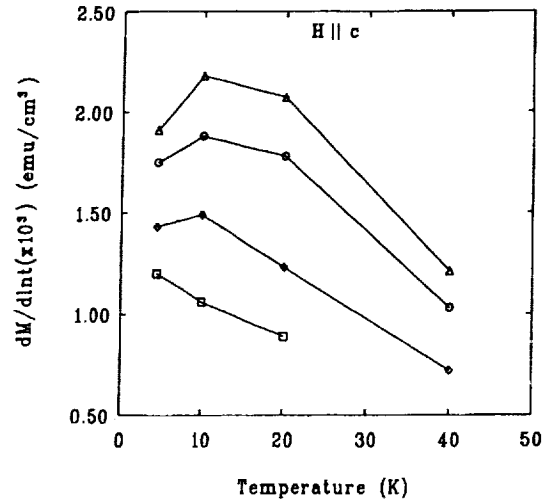
2. Experimental methods

The $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin film, approximately $0.3 \mu\text{m}$ thick, was deposited by the pulsed laser ablation technique onto a heated (100) LaAlO_3 substrate. The film texture was determined by the standard θ - 2θ scan and rocking curve, and shows that the grains are preferentially aligned with their c -axis along the plane normal. The film has a smooth surface and the average grain size, determined by scanning electron microscopy, is approximately $0.25 \mu\text{m}$. The superconducting transition temperature $T_c(0)$ determined by the standard four probe d.c. technique is 88.5 K with a transition width of *ca.* 1.0 K .

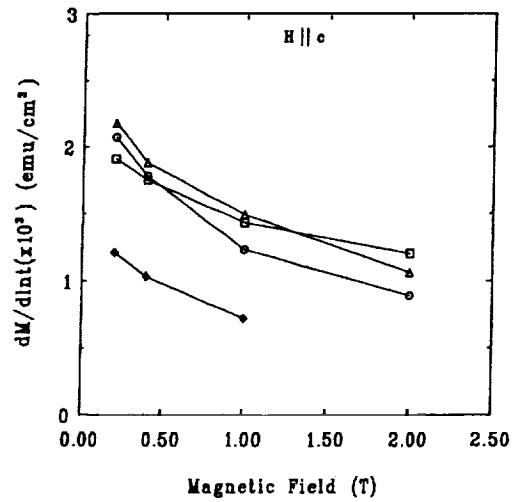
The magnetization and magnetic flux relaxation was studied using a commercial SQUID magnetometer. Hysteresis loops were made at various temperatures for $H \parallel c$, from which the critical current J_c was determined. The values at 4.2 K and 77 K were $2.4 \times 10^7 \text{ A cm}^{-2}$ and $1.0 \times 10^6 \text{ A cm}^{-2}$ respectively. Such values are typical of high quality films. The data collection procedure is described in detail in ref. 3. The diamagnetic transition temperature determined from the field-cooled magnetization of 2 mT applied along the c -axis was found to be 88.5 K and is the same as that determined by the electrical transport method.

3. Results

The relaxation of the screening-current-induced magnetization at $H = 0.2 \text{ T}$, 0.4 T , 1.0 T and 2.0 T applied perpendicular to the orthorhombic ab plane of the crystals was studied in the temperature range $0.05 T_c < T < 0.45 T_c$. The magnetization was found to relax logarithmically with t up to 10^4 s at all the temperatures and fields studied. The relaxation rate $dM/d \ln t$ is shown in Figs. 1(a) and 1(b) as a function of T and H respectively. It may be observed in Fig. 1(a) that $dM/d \ln t$ exhibits a maximum at approximately 10 K . Such behavior has been observed in grain aligned powdered $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ specimens [2] and attributed to partial flux penetration when $H < H^*$, where H^* is the field at which the magnetization reaches the maximum at a given temperature. In our relaxation measurements, however, $H > 2.5H^*$ for all temperatures and fields used. We therefore believe the films are fully penetrated by the flux and that the films are in the critical state. The relaxation rate $dM/d \ln t$ normalized by the initial magnetization M_0 at t_0 eliminates the uncertainties associated with the determination of the demagnetization



(a)



(b)

Fig. 1. The logarithmic relaxation rate $dM/d \ln t$ of the screening current induced magnetization: (a) As a function of temperature T at various fields Δ , 0.2 T ; \circ , 0.4 T ; \diamond , 1.0 T ; \square , 2.0 T ; (b) as a function of external field H at various temperatures \square , 4.2 K ; Δ , 10 K ; \circ , 20 K ; \diamond , 40 K . The lines connecting the data points are aids to the eye.

factor, and allows the possible determination of the pinning energy U_0 . In analyzing the results of the present work, t_0 is taken to be 10^3 s so that the relaxation is in the logarithmic regime. Figures 2(a) and 2(b) show the normalized relaxation rate $1/M_0 dM/d \ln t \equiv S$ as functions of T and H respectively.

4. Discussion

In a typical relaxation measurement, the net flux pinning energy U is zero at $t = 0$ and increases rapidly with t . The time dependence of U is implicitly contained

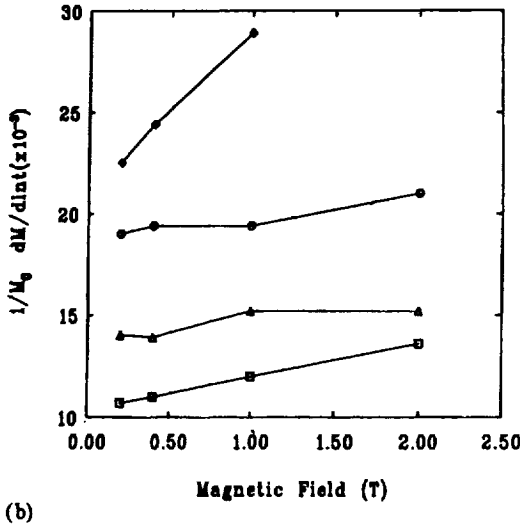
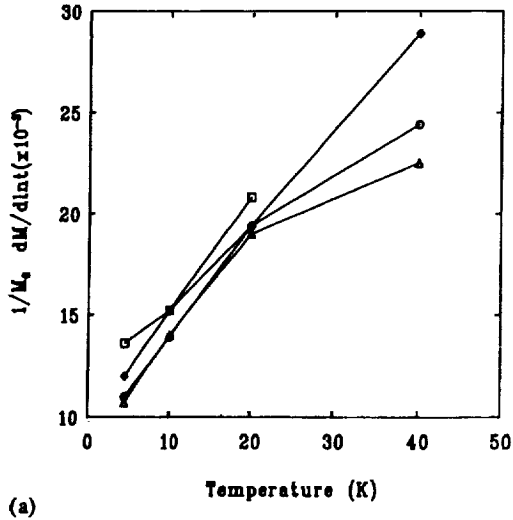


Fig. 2. The relaxation rate normalized by the initial magnetization M_0 : (a) as a function of temperature T ; (b) as a function of external field H . See caption of Fig. 1 notation.

within the screening current J which equals J_c at $t = 0$. The driving force for the motion of these flux lines is a combination of flux line interaction, thermal activation and the flux line gradient (Lorentz force). In the conventional thermally activated flux motion model, U is assumed to be a linear function of J and to have a depth U_0 when $J = 0$.

This leads to the classical relation [4, 5, 9] for $t \gg t_{\text{hop}}$,

$$M(t) = M(0)[1 - \{kT/U_0\} \ln(t/t_{\text{hop}})] \quad (1)$$

The relaxation rate normalized by the initial magnetization can be obtained from the above relation and is given as

$$1/M_0 dM/d \ln t \equiv S = -kT/[U_0 - kT \ln(t_0/t_{\text{hop}})] \quad (2)$$

According to the above relation, S is a linear function

of T when $U_0 \gg kT \ln(t_0/t_{\text{hop}})$. Since U_0 must go to zero as $T \rightarrow T_c$, it also predicts a divergence or an upward curvature for the S curve as T increases. From Fig. 2(a) it can be seen that S is approximately linear in T for $T \leq 20$ K, and takes on a weaker temperature dependence for higher temperatures. This type of behavior has been observed before in magnetization studies on single crystals, aligned powders and thin films [1-3]. The flux pinning energy U_0 obtained from eqn. (2) by considering t_{hop} to be typically 10^{-8} s is shown in Fig. 3(a). At every field U_0 is an increasing, convex function of temperature. The observation that U_0 is an increasing function of temperature rather than a decreasing function of temperature has led Hagen and Griessen [14] to propose that a distribution of U_0 values exists in these materials.

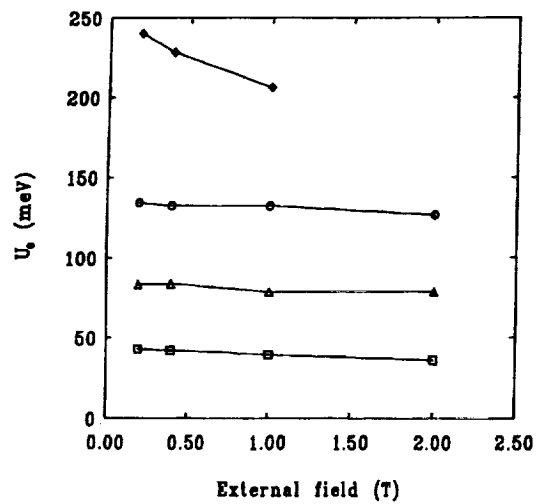
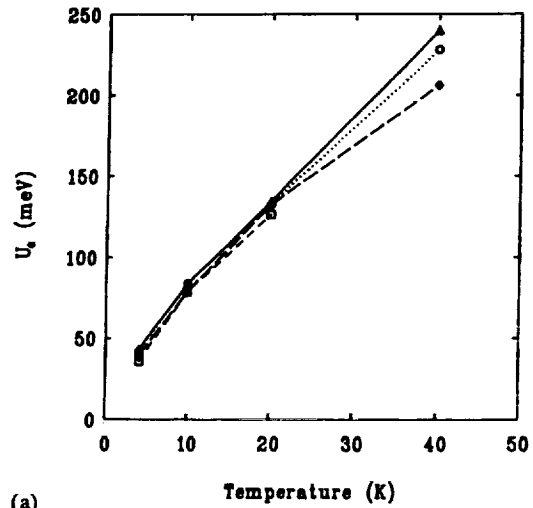


Fig. 3. The flux pinning energy U_0 obtained using eqn. (2) shown as a function of temperature (a) and external field (b). See caption of Fig. 1 notation.

In the vortex glass state, dissipation from the formation of vortex loops results in a magnetic relaxation of the form [12]

$$M(t) = M(0)[1 + (\mu kT/U_0) \ln(t/t_{\text{hop}})]^{-1/\mu} \quad (3)$$

where μ is the glass exponent. For times which are short compared with t_{cr} and where $\mu = 1$, eqn. (3) becomes equivalent to eqn. (1) and the resulting S is given by eqn. (2). In other words, at low temperatures the predictions of the activated flux creep and vortex glass are identical and in qualitative agreement with the data of Fig. 2(a). At times which are long compared with t_{cr} (or equivalently at higher temperatures) the vortex glass model predicts that $M \propto [\ln(t)]^{-1}$ and S takes on a temperature independent value given by $S = -[\ln(t/t_{\text{hop}})]$. Malozemoff and Fisher [12] argue that the nearly constant value of S reported in the literature is a consequence of the logarithmic dependence of S on the observation time. The data of Fig. 2(a) show that our values of S fall within the range of values observed by Malozemoff and Fisher, and that S tends to saturate for all fields as the temperature is increased. The latter fact is in qualitative agreement with the vortex glass model. While we have qualitative agreement with the temperature dependence predicted by the vortex glass model we do not observe the concomitant $[\ln(t)]^{-1}$ relaxation. Instead the magnetization decays as $\ln(t)$ at all temperatures used in this study.

Recently, substantial magnetic relaxation has been reported at temperatures as low as 0.1 K in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ grain aligned powder [15] and at 1.6 K in c -axis aligned thin film [16] for H applied along the c -axis. In the present work, however, 4.2 K was the lowest T at which the relaxation behavior was studied. In the low temperature limit both the thermally activated creep and the vortex glass models predict (eqn. (2)) that S vanishes as $T \rightarrow 0$. Extrapolation of the data (Fig. 2(a)) to $T = 0$ results in a non-zero S , in contradiction to both models. This behavior has also been observed in molybdenum disulfides for $T < 0.2 T_c$ by Mitin [13]. He has proposed that the observed relaxation results from *quantum tunneling or hopping* of the flux line segments across the potential barrier separating two pinning centers, and is athermal in nature. The hopping time for this process was estimated to be *ca.* 10^{-12} s, which is comparable to t_{hop} used in the present analysis. This phenomenon is similar in principle to the electron transport mechanism in disordered semiconductors [17]—quantum tunneling crossing over to thermal activation as T increases and domination of the highest rate process at any given T . At present there is no single model incorporating both these processes.

The magnetic field H dependence of U_0 is shown in Fig. 3(b). As can be seen from this figure, the

H dependence of U_0 changes continuously as T is increased. From this figure we observe that U_0 is weakly field dependent for $T \leq 20$ K. At all temperatures however, U_0 is observed to decrease with increasing field. On the other hand Xu *et al.* [2] observed U to increase with increasing fields for the same range of temperatures used in this experiment. Recent studies of the field dependence of U_0 in grain aligned [18] and single crystal [19] $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ have shown U_0 to increase with increasing fields at low temperatures and to decrease with increasing H for T near T_{irr} , where T_{irr} is the irreversibility temperature. This has been attributed to the variation of pinning strength, creation of field induced pinning centers and granularity. The disparity of the various field dependencies clearly shows that H dependence of U_0 is also not completely understood at present.

5. Conclusions

The temperature and magnetic field dependence of the flux pinning energy in a c -axis oriented $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin film have been investigated. The observed linear temperature dependence of S at low temperatures is consistent for both the thermally activated flux creep and the vortex glass models. The observed non-linear temperature dependence of S for $T \geq 20$ K is in agreement with the vortex glass model, but we do not observe the expected $[\ln(t)]^{-1}$ decay as predicted in this model. The behavior in the $T = 0$ limit, however, cannot be understood in terms of either model. At present one must resort to considering an athermal flux line tunneling mechanism for motion at $T = 0$. It therefore appears that no single model adequately describes the temperature, time, and field dependence of the magnetic relaxation in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ thin films.

Acknowledgments

S.V. wishes to acknowledge the U.S. National Research Council for an Associateship at NASA Lewis Research Center and M.A.S. acknowledges NASA for support through grant no. NAG-440751.

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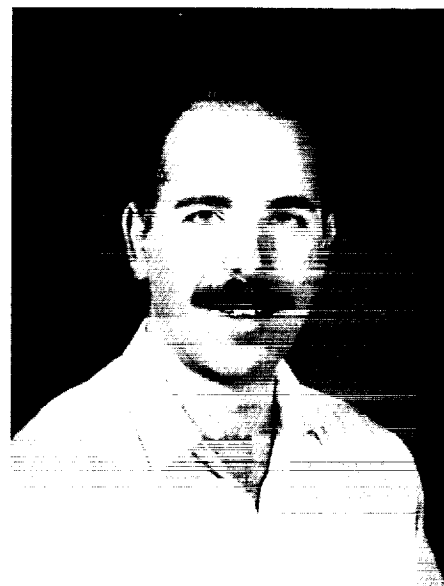
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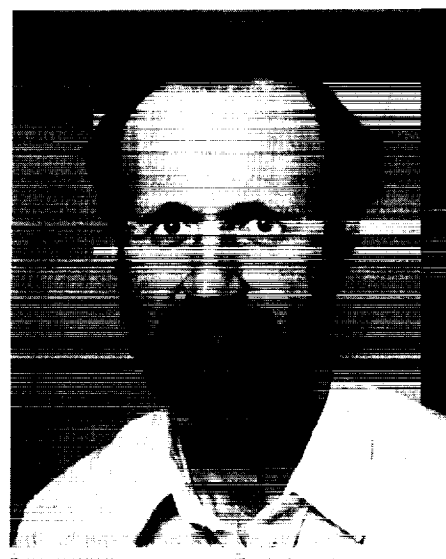
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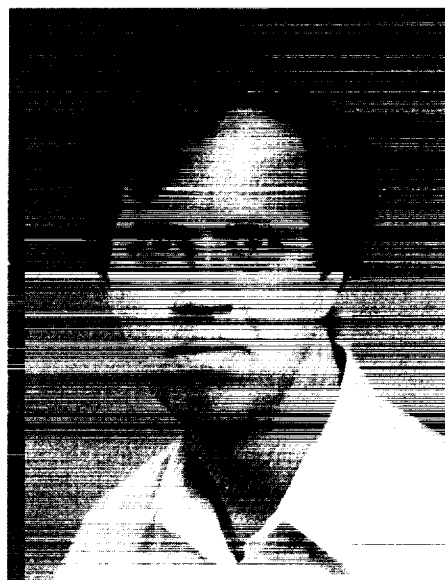
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